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TITLE: Materials Research Society Symposium Proceedings; Volume 720. Materials Issues for Tunable RF and Microwave Devices III Held in San Francisco, California on April 2-3, 2002

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Influence of the Laser Repetition Rate on Crystalline Structure, Composition, and Magnetic Properties of Laser Deposited $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ Films

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ABSTRACT

$Y_3Fe_5O_{12}$ (YIG) films have been deposited onto single crystal $Gd_3Ga_5O_{12}$ (GGG) substrates by pulsed laser deposition. For a given set of experimental parameters, the oxygen background pressure and substrate temperature were optimized to achieve the narrowest ferromagnetic resonance (FMR) lines. The repetition rate was then varied from 10 to 50 Hz. There is a clear transition from films with low saturation magnetization $4\pi M_S \approx 300$ Gs, high coercive fields $H_C > 20$ Oe, and broad FMR lines $\Delta H > 100$ Oe to films with $4\pi M_S > 1400$ Gs, $H_C < 10$ Oe, and $\Delta H \leq 10$ Oe. This crossover occurs when the laser repetition rate is changed from 20 to 30 Hz. No significant differences could be detected in any of the other investigated properties: crystalline structure, cation concentration ratio, and surface roughness do not depend on the repetition rate. Annealing experiments show that the films deposited at 10 and 20 Hz repetition rate are oxygen deficient. We loaded the film deposited at 20 Hz with oxygen, so that it reached the bulk value for $4\pi M_S$. The coercive field, however, remained large.

INTRODUCTION

$Y_3Fe_5O_{12}$ (YIG) films prepared by liquid phase epitaxy have attracted strong attention during the past decades. These films of the garnet structure have been grown with high perfection onto garnet single crystal $Gd_3Ga_5O_{12}$ (GGG) substrates, which have nearly zero lattice mismatch with the film. With sometimes sophisticated substitutions, garnet films have been proposed for two important applications in the past, as bubble memory films and for magnetostatic wave devices [1, 2]. In the past two decades, interest in their magneto-optical properties has grown [3–6].

In this paper, we investigate the magnetic, structural and compositional properties of laser deposited YIG films on GGG(111) substrates. Our investigations add information to prior publications about laser deposited YIG films [7, 8] on GGG substrates. We focus onto the interesting and technically important role of the laser repetition rate.

EXPERIMENTAL PROCEDURE

YIG films were grown onto GGG(111) single crystal substrates by the technique of pulsed laser deposition at 248 nm wavelength. Directly after preparation, the films were annealed in the deposition chamber at 700 mbar of oxygen pressure for 20 min. The target was ground before every new deposition.

The films were structurally characterized by three circle x-ray diffractometry (XRD). Film thicknesses were determined by profilometry over film edges and ranged between 400

and 600 nm. A qualitative picture of the film morphologies was obtained in an optical microscope. Film roughnesses were obtained in an atomic force microscope (AFM).

Rutherford Backscattering (RBS) allowed us to determine cation concentration ratios of the films and the target. We used He^4 ions at 1510 keV. The obtained backscattering spectra were fitted by the SIMNRA 4.0 simulation programme [9].

Magnetic moments and coercive fields were measured with a vibrating sample magnetometer. A broadband ferromagnetic resonance (FMR) spectrometer using a network analyzer as both source and receiver of the microwave radiation was used to determine position and width of the ferromagnetic resonance lines.

Further annealing was done in a separate oven under oxygen gas flow of 1000 cm^3/min at atmospheric pressure for 90 min at temperatures up to 950 °C.

RESULTS AND DISCUSSION

Keeping the laser energy fixed to 2.5 J/cm^2, the substrate-to-target distance to 4.5 cm, the laser repetition rate to 40 Hz, and the deposition time to 30 min (corresponding to 72000 pulses), we optimized the substrate temperature to 650 °C and the oxygen pressure to 0.034 mbar in order to achieve the narrowest FMR line of 6 Oe. According to our experience, the narrowest FMR line occurs under some specific angle, which is different for every film.

We found the best parameters of deposition at temperatures much lower than reported in Ref. [8], where the smallest FMR linewidth and the lowest coercive fields were found for substrate temperatures above 800 °C. In that investigation, however, the deposition rate (film thickness divided by total time of deposition) was twice as high as the one we used. We therefore concluded that the deposition rate plays a major role for the growth of laser deposited YIG films.

All films grew epitaxially, examples of a wide angle $\theta - 2\theta$ and a ϕ scan are shown in Figure 1. The (222) and (666) reflections are forbidden in the garnet structure and could stem from edges of the substrate or inhomogeneities of the film.

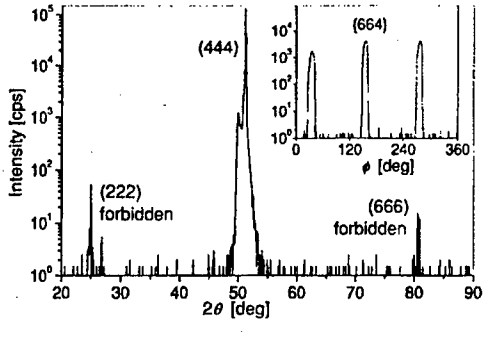

Figure 1. XRD patterns of a $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ film deposited at the laser repetition rate of 30 Hz. The main graph shows a wide angle $\theta - 2\theta$ scan. The inset shows a ϕ scan of the {664} oblique lattice planes.

It is impossible to see the main part of the film reflections in $\theta - 2\theta$ scans due to the extremely small lattice mismatch between YIG and GGG. This is demonstrated for films made with deposition rates from 10 to 50 Hz in figure 2. At deposition rates above 20 Hz, there are low intensity platforms to the left of the Bragg peaks. However, these platforms only have 1% of the peak height and do not show any peak structures.

Figure 3 shows the RBS spectrum of the film deposited at 30 Hz repetition rate. The edges in the spectrum correspond to the four heavy elements in the film and substrate. The oxygen edge is not resolved clearly. From carefully fitting the heights behind the Fe and Y edges, we determined the concentration ratio of Fe and Y irons (see inset of figure 3 for the fit). The cation concentration ratios in the other films and in the target were determined the same way. This analysis revealed slight Y deficiencies both in target and films (see table 1 for films, in the target $c_Y/c_{Fe} = 0.5$). The transfer from target to substrate was nearly stoichiometric for all films.

In transmitting light in the optical microscope, some differences between the films could be observed. The films deposited at the repetition rates of 30, 40, and 50 Hz look very similar: they show plain surfaces, with occasional voids. The film deposited at 20 Hz shows more and larger voids (around 10 μm in diameter), which possess irregular shapes. The film deposited at 10 Hz has even more voids, but they are smaller and circular. All films possess a large number of small dark spots, which are probably pits resulting from the small error in stoichiometry.

Film rms roughnesses averaged over areas of 20 μm^2 to 30 μm^2 are essentially the same and less than half the lattice parameter for all five films, see table 1.

The saturation magnetization is much higher for the three films deposited at the higher

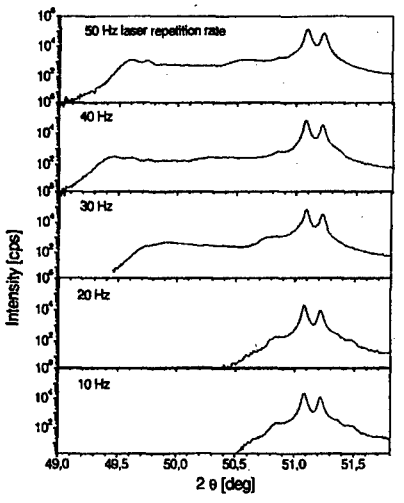

Figure 2. (444) reflections from $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ films deposited at identical conditions, except for the laser repetition rate, which is indicated for all spectra.

Table 1. Summary of the film properties at different repetition rates

	error	10 Hz	20 Hz	30 Hz	40 Hz	50 Hz
XRD: plateau left of (444)		no	no	yes	yes	yes
concentration c_Y/c_{Fe}	±0.01	0.49	0.50	0.56	0.47	0.52
rms roughness [nm]	±0.05	0.47	0.37	0.47	0.45	0.35
coercive field H_C [Oe]	±0.2	45	20	0.4	9	1.3
sat. magn. $4\pi M_S$ [Gs]	15%	310	250	1950	1590	1450
dark/light brownish color		dark	dark	light	light	light

repetition rates (table 1). Look at figure 4 to compare hysteresis loops (as prepared) of the films deposited at 20 and 30 Hz. The main error in determining the saturation magnetization comes from measuring sample volumes and can reach as much as 15%.

Figure 5 depicts the changes in coercive field and saturation magnetization as a function of the repetition rate. The coercive field H_C for 40 Hz is larger than for 30 and 50 Hz, which could be due to the better cation concentration ratio of the films deposited at 30 and 50 Hz.

The films also differ in color: The two films deposited at 10 and 20 Hz look dark brownish whereas the other three films look light brownish.

Considering the results described above, it seems likely that the differences in magnetic hysteresis loops are caused by differences in the oxygen contents of the samples. This hypothesis was tested by simultaneously annealing a 'bad' sample (deposited at 20 Hz repetition rate) and a 'good' sample (deposited at 30 Hz) for 90 min in 1 atm of oxygen at the temperatures of 500 °C, 650 °C, 800 °C, and 950 °C. Magnetic hysteresis loops were measured for both samples after every annealing step. The hysteresis loops of the good sample did not show any changes, but the saturation magnetization of the bad sample increased significantly and reached the same value as in the good sample (within the 15% error margin). The coercive field of the bad sample increased through annealing at 650 °C and then decreased

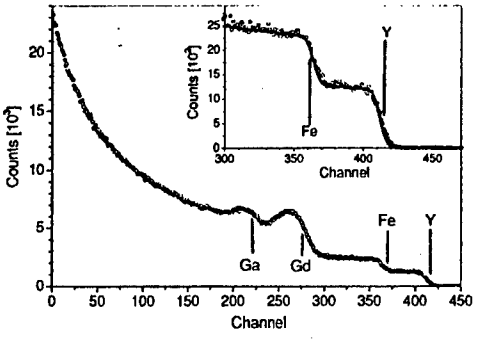

Figure 3. RBS spectrum of a $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ film deposited at the repetition rate of 30 Hz. The inset shows the fit of the film edges.

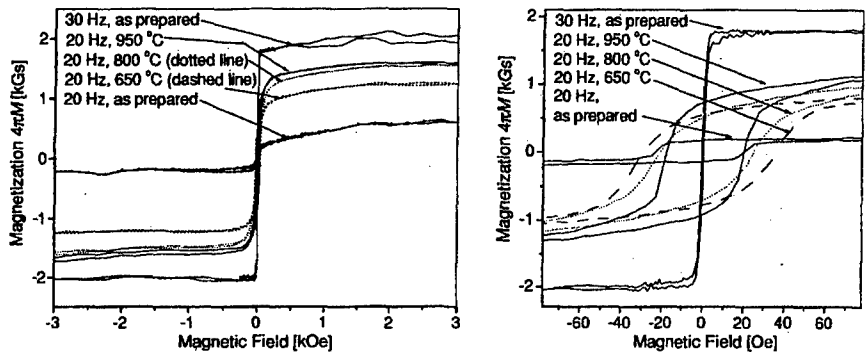

Figure 4. Magnetic hysteresis loops of two laser deposited $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ films recorded with the magnetic field parallel to the film plane. The films were deposited at the deposition rates indicated in the figure. After preparation they were subjected to postannealing in oxygen atmosphere for 90 min at the temperatures given in the left figure. The right figure shows the region of low magnetic fields in magnification.

again to approximately its original value. See figure 4 for hysteresis loops recorded with the magnetic field parallel to the film plane. The color of the bad film became lighter with every annealing step until both films looked the same after annealing at 800 °C.

The magnetic superexchange coupling in YIG is mediated by oxygen ions. Oxygen deficiency is therefore expected to result in reduced saturation magnetization. By increasing the oxygen content in the bad sample through annealing in oxygen atmosphere, the saturation magnetization was increased significantly.

In order to maintain charge neutrality at oxygen deficiency, a fraction of the metallic cations will change their valence states. Mixed valence states allow for charge transitions

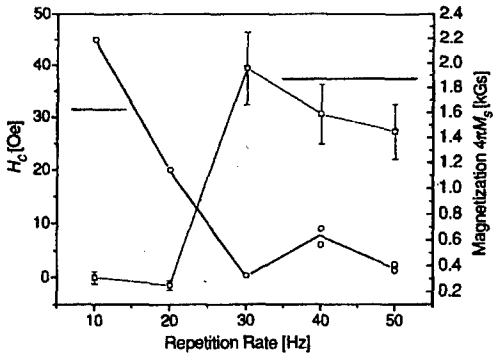

Figure 5. Coercive fields and saturation magnetizations for $Y_3Fe_5O_{12}/Gd_3Ga_5O_{12}(111)$ films deposited at different deposition rates.

and increase the optical absorption in the sample. The bad sample was therefore of a dark color directly after preparation, but its optical absorption was reduced through the annealing experiments.

One question remains, and this is *why* the two bad films possess oxygen deficiencies and the three good films do not. Most plausible is that oxygen left the growing film during the deposition process. There could be a change from films with microstructures permitting fast diffusion of oxygen to films with slower channels of oxygen diffusion as the repetition rate is increased from 20 to 30 Hz. Such a change in growth mode can occur abruptly when deposition parameters are changed over certain threshold values. The fact that the bad films possess larger coercive fields (even after annealing) supports the supposition that they are different from the good films with respect to microstructure.

CONCLUSIONS

The laser repetition rate is a very important parameter for the pulsed laser deposition of thin epitaxial YIG films. The search for films with narrowest FMR lines leads to different sets of experimental parameters, if the search is done for different fixed deposition rates. In our investigations, by changing the deposition rate and keeping all other parameters fixed, we observed a sharp transition in magnetic properties when the repetition rate was changed from 20 to 30 Hz. The reason is that films with low deposition rates are oxygen deficient.

One of our best films showed the following parameters: rms surface roughness is smaller than 1/3 of the lattice constant, saturation magnetization $4\pi M_S \approx 1950$ Gs is about the same as in crystalline bulk samples, the magnetic coercive field H_C is below 1 Oe, and the smallest FMR linewidth is $\Delta H \leq 10$ Oe. These values confirm that pulsed laser deposition is an interesting alternative to prepare high quality epitaxial YIG films.

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